LONG AND SHORT INTERVALS OF DERMAL EXPOSURE OF PEACH HARVESTERS TO FOLIAR AZINPHOS-METHYL RESIDUES

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SUMMARY

A dermal monitoring study of peach harvesters exposed to azinphos-methyl residues was conducted at two sites to determine the relationship between dermal exposure and time or production. Workers wore two long-sleeved knit t-shirts for each monitoring interval and provided a hand residue sample. Dislodgeable foliar samples were also collected. The highest correlation at both sites was found between production and inner shirts ($p<0.01$), indicating that two layers of dosimetry clothing provide a more accurate measure of dermal exposure than a single layer or patches. Residue acquisition on inner shirts became constant after three hours, indicating that exposure estimates from shorter intervals would over-estimate exposure. Transfer factors (cm^2/hr) were developed from dermal and dislodgeable data and were consistent with those suggested by previous investigators.

INTRODUCTION

Historically, CDFA has conducted dermal monitoring of workers over an entire workday to enable an estimate of total dermal exposure. However, recent data indicate that pesticide residues may not transfer to a worker in a linear manner with time or production, but may load on the sampling media during the initial hours (Fenske et al. 1989; Zweig et al. 1985; Davis et al. 1983; Noel et al. 1983). If monitoring media exhibit uneven acquisition or loading effects, then extrapolating from residues transferred over a one- or two-hour exposure to an eight-hour exposure may over-estimate dermal exposure. Similarly, monitoring a worker for eight hours and assuming the resultant dermal residues were transferred linearly over the workday may underestimate the residues available for dermal absorption during the early portion of the day. It is important to measure the rate of pesticide residue transfer from crop to monitoring media in order to make the best possible exposure assessments for incorporation into risk assessments and for development of risk mitigation measures.

In August and September, 1989, the California Department of Food and Agriculture (CDFA), Worker Health and Safety Branch, conducted a dermal monitoring study of peach harvesters exposed to azinphos-methyl residues at two sites in Sacramento and Stanislaus Counties to investigate the relationship between dermal exposure and time or production. Exposure was measured with two long-sleeved knit undershirts worn for the duration of each monitoring period and a hand residue sample at the end of the monitoring period. Shirts were employed instead of traditional gauze pad dosimetry (Durham and Wolfe 1962) as they offer greater sample integrity than gauze pads and allow an estimate of upper body dermal exposure without further extrapolations. Dislodgeable foliar residue samples were also collected.

In addition, residue transfer factors (cm^2/hr) were developed from the ratio of hourly dermal exposure residues to dislodgeable foliar residues to describe the rate of pesticide residue transfer from crop to worker (Popendorf and Leffingwell 1982; Nigg et al. 1984; Zweig et al. 1983; Zweig et al. 1985). These transfer factors are compared to those calculated in both previous Branch studies and by other investigators.

MATERIALS AND METHODS

Twenty-nine peach harvesters in Sutter County and 12 in Stanislaus County were monitored for dermal exposure to azinphos-methyl during one workday. All study participants were male. The orchards in Sutter County were treated once 50 days prior to the study at the rate of 1.5 lb. active ingredient (a.i.) per acre, applied in 100 gallons of water. The orchards in Stanislaus County were treated once 74 days prior to the study at the rate of 0.75 lb a.i. per acre, applied in 100 gallons of water. The California reentry interval for peach orchards treated with azinphos-methyl is 14 days (State of California, Department of Food and Agriculture, 1989). All chemical analyses were conducted by CDFA Chemistry Services Laboratory, Worker Health and Safety Branch, Sacramento.

Dislodgeable Foliar Residue Monitoring

The orchards were sampled for dislodgeable foliar residue (DFR; Gunther et al. 1973). Each sample consisted of sixty leaf disks, 1.78 cm in diameter, cut with a Birkestrand punch. Each sample was taken from 10 trees at a height of 5-6 feet. Three locations in each orchard were sampled. Sample jars were sealed with aluminum foil, capped and kept on ice for shipment to the laboratory for analysis.

Dermal Monitoring

The workers at both sites wore two long-sleeved, 100% cotton, knit undershirts in place of regular work shirts for each monitoring interval. At the end of the monitoring interval, each worker provided a hand residue sample. At the Sutter site, these were obtained by the workers wiping their hands with two pre-moistened disposeable wipes (Chubs^R brand, 5.75 x 8 inches). All wipe samples were stored in four-ounce glass jars, which were covered with aluminum foil and capped. In Stanislaus, hand residues were obtained by a wash. Each worker washed his hands for one minute in 500 ml of 1% sodium dioctyl sulfosuccinate (Sur-Ten^R) contained in a one-gallon polyethylene bag. All handwash samples were poured into Nalgene^R bottles. After providing a hand residue sample, the workers removed both dosimetry shirts and placed each one in a separate one-gallon polyethylene bag with a single track seal. All dermal samples were stored on dry ice until extraction.

Sample Analysis

All samples were analyzed for the presence of azinphos-methyl and its oxon. The shirts, wipes and hand washes were extracted with ethyl acetate and dried with anhydrous sodium sulfate. The foliar samples were shaken three times each with 50 ml sodium dioctyl sulfosuccinate solution. The combined DFR extracts were extracted three times using 50 ml ethyl acetate which was then dried by the addition of sodium sulfate. The samples were analyzed by gas liquid chromatography on a Hewlett-Packard 5880A chromatograph equipped with a nitrogen-phosphorus detector. The chromatographic conditions were: column, 10m x 0.52 mm HP 50% phenyl methyl silicone; carrier gas (He), 20 ml/min; H_2, 2 ml/min; air, 90 ml/min; injector temperature, 275° C; oven temperature, 235° C isothermal. The retention time of azinphos-methyl using the above conditions was 6.49 minutes and for the oxon, 5.32 minutes. Minimum detectable levels for azinphos-methyl in micrograms per sample were 0.25, 5.0, 1.0 and 1.0 for the leaf disks, shirts, wipes and washes, respectively. Minimum detectable levels for the oxon in micrograms per sample were 0.5, 10.0, 1.0 and 2.0 for the leaf disks, shirts, wipes and washes, respectively. Results reflect the sum of azinphos-methyl and oxon residues for each sample type.

Data Analysis

Least squares linear regression was performed on the dermal media residues vs time and production. Critical values of r were determined by table for a two-tailed test at $p<0.10$. T-tests were performed on the means of the dermal exposures per bin vs work interval for the Sutter site to determine if residue loading occurred. The level of statistical significance selected was $p<0.10$.

RESULTS

Dislodgeable Foliar Residues (DFR)

The mean azinphos-methyl (AM) DFRs were 0.37 ± 0.06 ug/cm^2 (n=3) and 1.0 ± 0.08 ug/cm^2 (n=3), respectively, for the test orchards in Sutter and Stanislaus counties. No oxon was detected on any of the foliar samples (MDL = 0.0025 ug/cm^2). Although AM was applied to the Sutter orchard at 2 times the rate applied in Stanislaus and 50 days prior to the study compared to 74 days in Stanislaus, mean residues in Stanislaus were nearly three times those in Sutter. The orchard in Sutter was younger, bushier, more open and had greater light penetration than the Stanislaus orchard, which could have resulted in a greater rate of photodegradation of the residues. The mean DFR for each orchard was employed in calculating transfer factors to describe the rate of AM residue transfer from peach foliage to harvester.

Dermal Monitoring

The mean values for AM plus AM oxon on the dermal monitoring media for each group of workers at each monitoring interval are in Table I. The overall means for the outer shirt residues were similar for the two sites with values of 17.5 mg and 16.7 mg for Sutter and Stanislaus, respectively. The means for the inner shirts and hands each differed by about two-fold between the two sites. The inner shirt means were higher in Sutter (9.0 mg compared to 3.8 mg in Stanislaus) while the hand residues were higher in Stanislaus (3.6 mg compared to 2.0 mg in Sutter). The oxon on the monitoring media averaged 5-15% of the AM levels. Because the oxon is approximately 30 times more toxic than the thion, this level could be significant in terms of risk (Popendorf and Leffingwell, 1982). However, the mechanism of oxon formation on clothing and skin in the absence of oxon presence on leaf surfaces remains unclear. Thus, the oxon levels were not reported separately for the dermal monitoring.

The correlation coefficients and significance levels for the various media vs production and time are presented in Table II. The best correlation at both sites was found between inner shirts and production (r = 0.78, 0.88 for Sutter and Stanislaus, respectively), and inner shirts and time worked (r = 0.75, 0.87 for Sutter and Stanislaus, respectively). Outer shirts, hands and dermal exposure (inner shirts + hand residues) vs time and production were less correlated. The correlation between dermal exposure and time or production was significant for both sites at the 0.01 level, due to the contribution of inner shirt values. Correlations of outer shirts and hands vs time or production were significant at the 0.01 level only for the Sutter site. Although r values for outer shirts and hands vs time or production were similar or higher at the Stanislaus site, the smaller sample population (n = 12 in Stanislaus, n = 50 in Sutter) restricted the significance level to 0.10 and 0.05 for the outer shirts and hands, respectively.

Comparisons of transfer factors from three studies are presented in Table III. Among the three studies which employed outer shirt and hand residues to estimate exposure, the DFR differs by three-fold (0.37-1.00 ug/cm^2), hourly exposure differs by nearly four-fold (2150 - 7800 ug/hr) and the calculated transfer factors differ by less than two-fold (6900 - 13,200 cm^2/hr). Among the four studies using inner shirt and hand residues to assess exposure, the DFR differs by more than 30-fold (0.03-1.00 ug/cm^2),

hourly exposure differs by about about 11-fold (250 - 2820 ug/hr) and the transfer factors differed by about three-fold (2800 - 7430 cm^2/hr).

The plots of the regressions of AM residues on the monitoring media vs production are given in Figures 1 and 2. Although there is greater than two-fold variation in the mean residues between the two sites (9.0 and 3.8 mg for Sutter and Stanislaus, respectively), the rates at which the inner shirts acquired residues were similar (slope = 1057 ug/bin and 1063 ug/bin for Sutter and Stanislaus, respectively). In contrast, the mean outer shirt residues are similar for both sites, but the rate at which the outer shirt collected residues was three-fold greater at Stanislaus (4.2 mg/bin, 1.5 mg/bin for Stanislaus and Sutter, respectively).

Individual worker production rates were not available for the Stanislaus site as four men picked into a bin rather than picking to individual bins as at the Sutter site. For the Sutter County site, the distribution of worker production rates is given in Figure 3. The plot shows a normal distribution about picking rates that vary by two-fold (range = 31 - 65 minutes/bin, with a mean and median of 44). The dermal exposure/bin vs time for the Sutter site is presented in Figure 4 and outer shirt residue/bin vs time is given in Figure 5. Figure 4 shows a significantly higher exposure/bin ($p<0.01$, 2.7 mg/bin/min) for the first interval compared to any of the succeeding three intervals (mean for last three intervals = 1.9 mg/bin/min). Figure 5 shows that the rate of acquisition of outer shirt residues/bin decreases constantly with time and never achieves equilibrium ($r = 0.66$, $p < 0.01$).

DISCUSSION AND CONCLUSIONS

Dermal Exposure

Estimating dermal exposure to pesticide residues using the traditional method of mounting gauze pads outside a worker's clothing can result in inaccuracies since residues must first be extrapolated from the pad area to the corresponding body surface area and then to the residues calculated to penetrate clothing and be available for absorption at the skin level. To overcome these sources of inaccuracy, a "double dosimetry" system of two long-sleeved t-shirts worn together was employed to measure both the outer and "skin level" exposure to AM residues. The residues on the two shirts were compared and a clothing penetration value obtained empirically. Using group means from Table I, measurements of both potential dermal exposure (outer shirt + hand residues) and dermal exposure (inner shirt + hand residues) were similar for the two sites.

Potential dermal exposure averaged 19.5 $\pm$ 3.9 mg in Sutter and 20.3 $\pm$ 7.7 mg in Stanislaus. Dermal exposure averaged 11.0 $\pm$ 3.7 mg in Sutter County and 7.3 $\pm$ 0.6 mg in Stanislaus County. The percent contribution of the inner shirt to dermal exposure was lower at each time interval for the Stanislaus site (51 $\pm$ 14%) than for the Sutter site (80 $\pm$ 5%) (Fig. 1-2).

Shirt Penetration

The mean outer shirt residues were similar for both sites (17.5 mg in Sutter, 16.7 mg in Stanislaus). Because the mean inner shirt residues were lower in Stanislaus (3.8 mg in Stanislaus, 9.0 mg in Sutter), the mean shirt penetration values [(inner shirt/inner + outer shirt residues) x 100] varied

by about 50% for the two sites (33% and 21% for Sutter and Stanislaus, respectively, Table II, Fig.1-2). This range is similar to the 25-47% of residues on clothing estimated to reach the skin by Popendorf et al. (1979), using inner and outer gauze pad dosimetry. CDFA currently employs a default value of 10% for clothing penetration (clothing protection) through normal work clothing (Thongsinthusak et al. 1990). Data from this study indicate that two long-sleeve shirt layers may provide 2-3 times less protection than this value for harvester exposure to AM residues. Since field observation has shown that normal work clothing for tree fruit harvesters consists of a long-sleeve button shirt worn over a short-sleeve undershirt, clothing protection may be even less, as the standard work clothing provides no second layer to intercept forearm residues. There was a significant relationship at the Sutter site between percent shirt penetration vs both production ($p<0.01$) and time ($p<0.02$).

Exposure vs Time or Production

In Stanislaus, three groups of four harvesters each were monitored for one interval of 1.3, 2.6 or 4 hours. In Sutter, three groups of seven harvesters each were monitored for two sequential intervals of 1.5 to 5.5 hours and one group of eight harvesters was monitored for the entire workday (7 hours) (Table 1). Data were analyzed to determine differences between the two sequential intervals in the amount of residues transferred to the dermal media. There was no statistical difference between the residue means in the paired groups 1.5 and 2 hr, 3 and 4 hour, and 5 and 5.5 hr, indicating the dermal residues were not dependent on the sequence of monitoring interval. Therefore, the results were grouped into four work periods for any further time/residue comparisons: three monitoring intervals, each composed of 14 subjects (1.5-2 hours, 3-4 hours, 5-5.5 hours) and the all-day group (7 hours), composed of eight subjects.

Regression analyses on the residues on the individual media vs time or production gave similar results (Table II). However, the correlation with production was slightly better than with time, and subsequent analyses were made with production. Since, at the Sutter site, the picking rate per worker had a normal distribution about rates which varied by two-fold (Figure 3), it is expected that dermal exposure is better correlated with production than with time. Previous investigators have found a higher correlation between exposure and production rate than between exposure and time worked (Fenske et al. 1989). Nigg et al. (1984) found significant correlations between production and the concentration of DDA metabolite in urine of citrus harvesters exposed to chlorobenzilate residues ($r = 0.71$) as well as between work rate and total body exposure ($r = 0.76$). The correlation of exposure with time worked was not investigated in Nigg's study as the monitoring period was for the 8-hour workday and no samples were collected at intermediate intervals. While time worked is easily documented, for a more complete understanding of worker exposure, it is important to document workers' daily production, when possible.

The highest correlation at both sites was found between production and the inner shirt (dermal) residues. The mean inner shirt residues differed by three-fold between sites but had similar rates of residue acquisition. Mean outer shirt residues were similar for the two sites, but had residue acquisition rates that varied by three-fold. Therefore, the outer shirt filters residues to the inner shirt (skin) at a constant rate, dependent on

pesticide residue and fabric characteristics, as previously reported by Davies et al. (1980). The three-fold greater rate of residue transfer to the outer shirt at the Stanislaus site is likely related to the greater DFR at that site (1.0 ug/cm^2 compared to 0.37 ug/cm^2 at Sutter).

Outer shirt residues were less correlated with production than were inner shirts. This suggests that errors may arise when calculating estimates of dermal exposure (skin level residues) from measurements of potential exposure (outer shirt or patch data) using an estimated value for clothing penetration. It appears that pesticide exposure monitoring studies yield more realistic estimates of dermal exposure when inner shirt data is used. Future studies could include the use of a woven, buttoned shirt as the outer shirt since observation has shown that this is the typical outer clothing for peach harvesters. Preliminary studies conducted by CDFA with a mannequin-type sampler show similar penetration values through woven and knit shirts (J. Sanborn, pers. comm.).

The hand residue values were higher for the Stanislaus site (3.6 $\pm$ 0.8 mg compared to 2 $\pm$ 0.7 mg in Sutter) and accounted for more than 50% of the dermal exposure. It is not clear whether the higher hand residue values were due to the higher DFR at the Stanislaus site or a greater efficiency in residue removal by hand washes compared to hand wipes. However, preliminary studies by CDFA have found wipes remove 62-82% of residues removed by hand washes (C. Blewett and F. Schneider, pers. comm.). The correlation of hand washes with production was better at the Stanislaus site than for hand wipes vs production at Sutter. Hand wipes offer advantages over hand washes in that they require less advance preparation, are readily available at retail outlets, sampling time is reduced thus creating less work task interruption, sampling media are lighter (wipes vs 500 ml water), have less potential for hydrolysis during storage and require less solvent for extraction. Davis et al. (1983) noted the disadvantages of hand washes and found that, in comparison, gloves over-estimate hand exposure. Later investigations by Fenske et al. (1989) confirmed this observation. Further studies will determine whether hand wipes and hand washes provide similar exposure values.

Residue Loading Effects

The payment of harvesters at Sutter by piecework provided an opportunity to investigate the individual variation in dermal media residue accumulation. Since inner shirts were more correlated with production than outer shirts, dermal exposure data from the Sutter site, normalized to exposure/bin, were analyzed to characterize residue equilibria on sampling media (Figure 4). Equilibrium in the rate of residue acquisition over time would be indicated by equal means for each of the four monitoring intervals. The mean for the first interval, 1.5-2 hours, showed a significantly higher (student's t-test, $p<0.01$) dermal exposure per bin (2.7 mg) than for any of the succeeding intervals. There was no difference among the means for the last three intervals. This 1.5-fold higher exposure/bin for a two-hour interval compared to longer intervals indicates that sampling media may have a higher affinity for AM residues early in the exposure (loading) and may require up to three hours to reach a residue equilibrium. It appears that a sampling interval of three hours would give a dermal exposure that could be extrapolated to longer time intervals without being skewed by the effects of residue loading seen in the first two hours. Conversely, extrapolating to an

8-hour dermal exposure from a sampling interval of less than three hours would be likely to over-estimate actual exposure by 50-60% (2.7 mg/bin for 2 hrs vs 1.8 mg/bin for 7 hrs). Data for all media from the Sutter site were then examined in terms of residues/bin vs time worked to determine if media loading occurred. Only outer shirts displayed a significant linear relationship (Figure 5), indicating that the rate of residue acquisition was not constant. Outer shirt residues/bin never achieved equilibrium over an exposure period as long as 8 hours and, in fact, decreased continuously with time. While Fig. 1 and 2 show an increase in total residue acquisition on outer shirts with increasing production, Fig. 5 indicates that outer shirts load during the initial exposure hours and the rate of residue accumulation per bin decreases with time. Therefore, estimates of dermal exposure from outer shirt/bin data at any time interval less than a full work period would be likely to over-estimate exposure.

Transfer Factors

Several investigators have developed a ratio of hourly dermal exposure to DFR [(ug/hr)/(ug/cm^2)] to describe the rate of transfer of foliar residues from crop to worker (cm^2/hr) (Popendorf and Leffingwell 1982; Nigg et al. 1984; Zweig et al. 1983; Zweig et al. 1985). CDFA has been investigating the variation of these transfer factors with both crop and work task. The transfer factors using the outer shirt plus hand residues are 13,200 cm^2/hr for the Sutter site and 7800 cm^2/hr for the Stanislaus site. Previous Branch studies using outer shirt and hand dosimetry (potential dermal exposure) to assess exposure to AM observed transfer factors for harvest of stone fruits of 6900 cm^2/hr (Schneider et al. 1990, HS-1532, DFR = 0.31). In this study, the transfer factor using the inner shirts plus hand residues (dermal exposure) was 7430 cm^2/hr for the Sutter site and 2850 cm^2/hr for the Stanislaus site. The transfer factors found in previous Branch studies of AM exposure using inner shirt plus hand dosimetry were 3050 cm^2/hr and 8300 cm^2/hr (Spencer et al. 1990, HS-1577, DFR = 0.03 and 0.63, respectively). DFR values among these three studies differed by as much as 30-fold while the transfer factors differ by only five-fold (Table III).

CDFA's calculated transfer factors are similar to the 10,000 cm^2/hr suggested by Nigg et al. (1984) for residue transfer to citrus harvesters. The relative consistency of transfer factors as compared to DFR values suggests that transfer factors may allow estimation of dermal exposure of harvesters in stone fruit. While transfer factors can be developed from either inner or outer shirt data, inner shirts provide a more accurate estimate of skin level exposure than do outer shirt data, which must be adjusted for estimated penetration. CDFA will continue to investigate the validity of transfer factors to estimate dermal exposure.

Table I
Mean Dermal Dosimetry Values by Site and Interval

Site	N	Hours/interval	mg (azinphos-methyl + oxon)/interval			
Sutter		First	Outer Shirt	Inner Shirt	Hands	% Shirt Penetration
	7	1.5	13.3 ± 2.9	5.6 ± 0.5	1.4 ± 0.6	30
	7	3	20.8 ± 0.5	7.1 ± 1.7	1.7 ± 0.6	25
	7	5	20.4 ± 7.2	13.9 ± 3.8	2.1 ± 0.6	41
	8	7	21.5 ± 9.7	12.5 ± 2.5	3.5 ± 1.5	37
		Second				
	7	2	13.1 ± 2.4	6.2 ± 7.0	2.3 ± 7.3	32
	7	4	16.0 ± 3.0	6.8 ± 1.1	1.8 ± 6.0	30
	7	5.5	17.1 ± 7.1	10.6 ± 3.2	1.5 ± 8.2	38
Mean ± SD		4	17.5 ± 3.5	9.0 ± 3.3	2.0 ± 0.7	33 ± 6
Stanislaus						
	4	1.3	9.3 ± 2.8	5.2 ± 2.0	2.7 ± 1.0	36
	4	2.6	23.1 ± 6.9	2.7 ± 3.1	4.0 ± 2.8	10
	4	4	17.7 ± 7.3	3.4 ± 1.1	4.0 ± 6.4	16
Mean ± SD		2.6	16.7 ± 7.0	3.8 ± 1.3	3.6 ± 0.8	21 ± 14

Table II

r Values for Dermal Exposure Correlated with Time or Production

	Sutter		Stanislaus	
Dermal Media	r	p value	r	p value
Dermal Exposure\a				
per minute	0.46	p< 0.01	0.80	p< 0.01
per bin	0.60	p< 0.01	0.81	p< 0.01
Inner Shirts				
per minute	0.75	p< 0.01	0.87	p< 0.01
per bin	0.78	p< 0.01	0.88	p< 0.01
Outer Shirts				
per minute	0.53	p< 0.01	0.45	p< 0.10
per bin	0.57	p< 0.01	0.48	p< 0.10
Hands				
per minute	0.45	p< 0.01	0.58	p< 0.05
per bin	0.41	p< 0.01	0.60	p< 0.05
% Shirt Penetration				
per minute	0.36	p< 0.02	0.14	p> 0.10
per bin	0.40	p< 0.01	0.14	p> 0.10
n	50	50	12	12

\a Inner shirt + hand residues

Table III
Comparisons of Azinphos-methyl Transfer Factors (TF[a]) in Stone Fruits

County	Shirt		Hand	Exposure	DFR	TF
	Outer	Inner		(ug/hr)	(ug/cm^2)	(cm^2/hr)
Fresno[b]	x		x	2150	0.31	6900
Sutter[c]	x		x	4875	0.37	13,200
Stanislaus[c]	x		x	7800	1.00	7800
Sutter[d]		x	x	1950	0.64	3050
Stanislaus[d]		x	x	250	0.03	8300
Sutter[c]		x	x	2750	0.37	7430
Stanislaus[c]		x	x	2820	1.00	2850

a TF = Dermal Exposure/DFR
= (ug/hr)/(ug/cm^2)
= cm^2/hr
b HS-1532
c present study
d HS-1577

Fig. 1 Dermal Monitoring Residues vs Production, Sutter Co., 1989

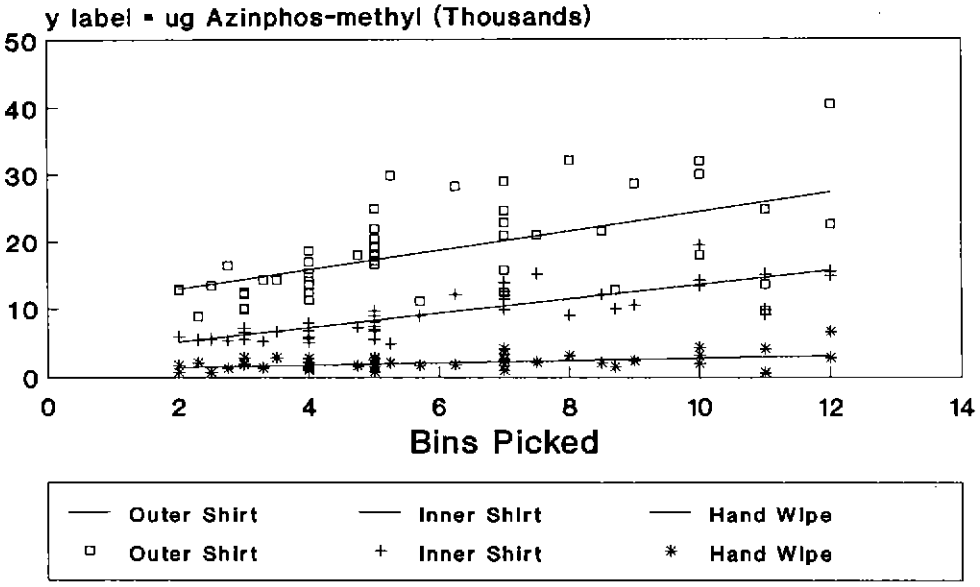


Fig. 2 Dermal Monitoring Residues vs Production, Stanislaus Co., 1989

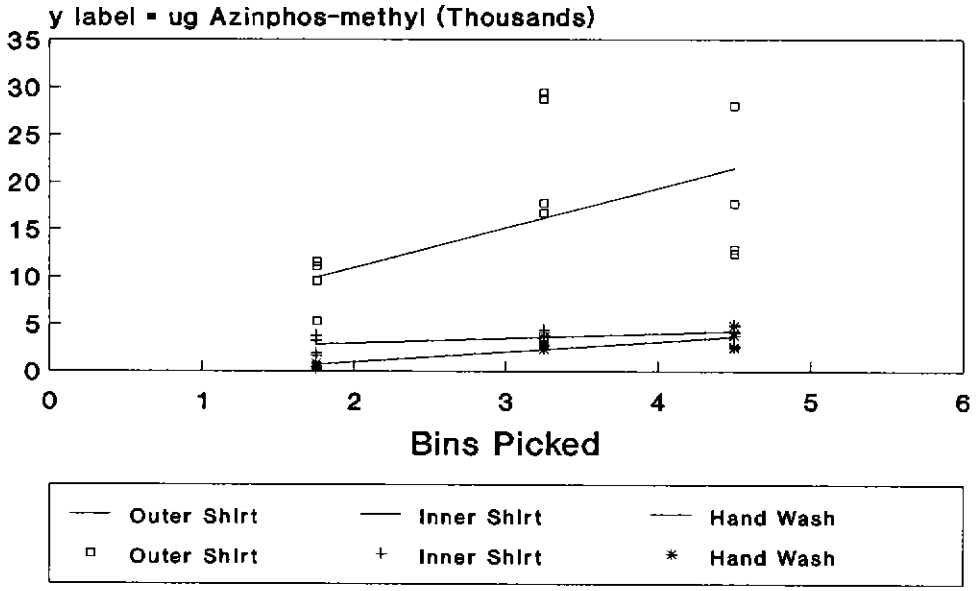


Fig. 3 Production Rate Distribution Peach Harvesters, Sutter Co., 1989

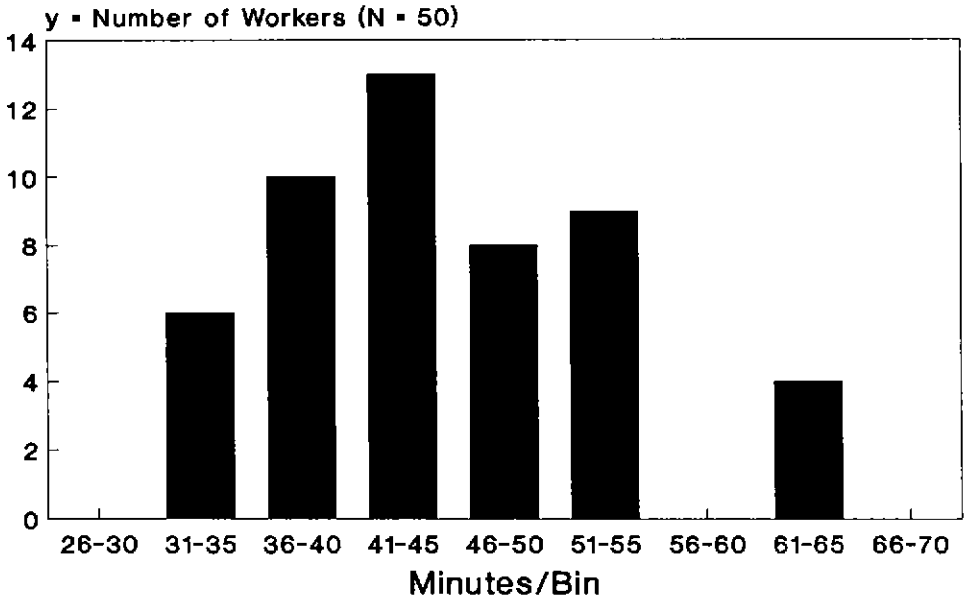


Fig. 4 Dermal Exposure/Bin vs Time Peach Harvesters, Sutter Co., 1989

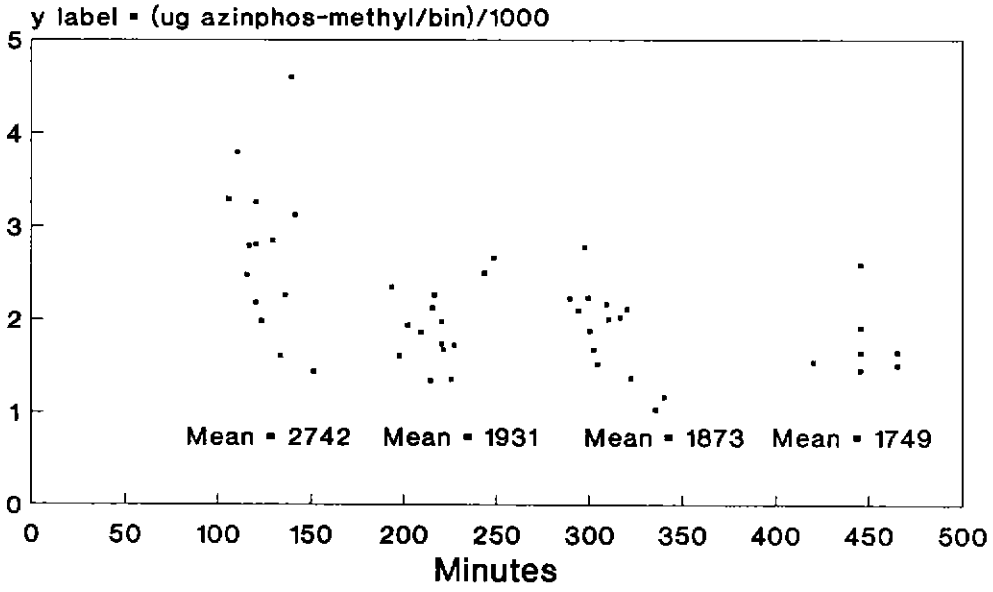
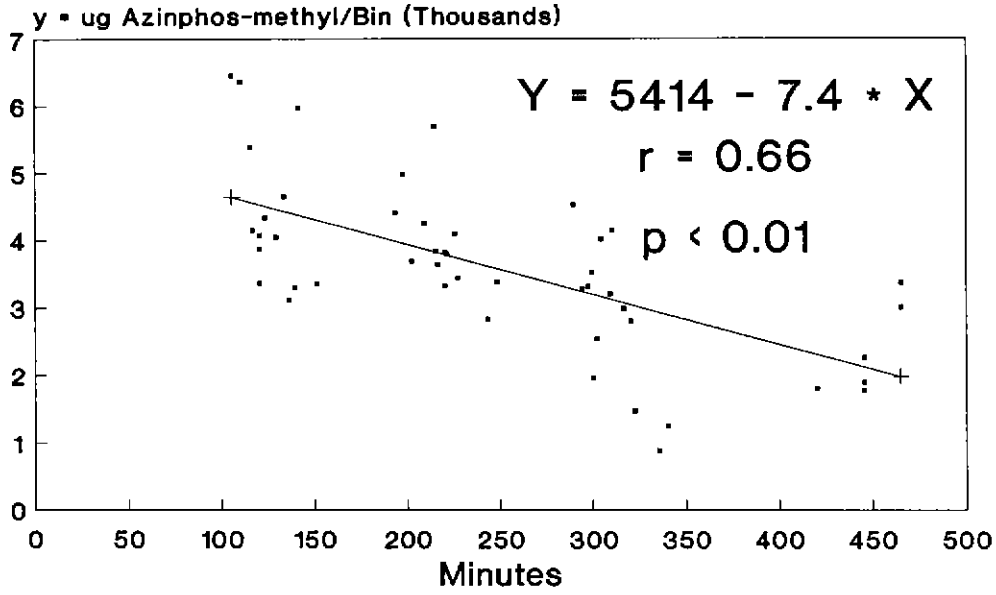


Fig. 5 Outer Shirt Exposure/Bin vs Time Peach Harvesters, Sutter Co., 1989



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